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Liposome-based SERS Sensor system for double readout of intracellular Iron concentrations and pH changes

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Liposome-based SERS Sensor system for double readout of intracellular Iron concentrations and pH changes

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ABSTRACT: Nanoscopic sensor systems based on Fluorescence and SERS microscopy are commonly employed for the detection of biologically relevant ions in cells and remain promising tools for advancing medical diagnostics and enhancing the understanding of ion-related processes. While these techniques allow spatially and temporally resolved read out of even low ion concentrations, a common barrier encountered in developing robust optical ion sensors for applications in diagnostics or bio-sensing is crosstalk with the pH of the sample. Often, the reporter molecules employed for binding the ions of interest and enabling readout via fluorescence or SERS microscopy are highly sensitive to the pH prevalent in biological samples. Among the biologically and medically relevant ions, iron stands out as being one of the most abundant and important ions. Given the rising interest in Ferroptosis, the accurate, localized and time resolved measurement of intracellular iron concentrations has become crucial in deepening the understanding of this process.

Here, we present an iron and pH double readout sensor system, enabled by a very versatile sensor system that is based on the amine-promoted decoration of polystyrene beads with silver nanoparticles and their subsequent encapsulation within liposomes. The iron sensor utilizes hydrophobically entrapped phenanthroline within the liposomal bilayer, while the pH sensor utilizes covalently bound 4-mercaptobenzoic acid. The liposome coating provides biocompatibility, hydrophobic entrapment, and tuneable membrane properties, for example through the choice of lipids with specific phase transition temperatures or by incorporating unsaturated fatty acids such as oleic acid to modulate membrane permeability. Combining the two SERS sensors allows for the simultaneous differentiation between false positive iron concentration readouts induced by pH changes and actual positive readouts caused by changes in iron concentration. Furthermore, the combination of microscopic images, pH readout, and liposomal encapsulation could help refine the accurate localization of the sensor system, distinguishing between acidic endosomes and the neutral cytoplasm. In processes such as ferroptosis, the simultaneous read out of pH changes and the iron concentration in lysosomes could uncover new correlations and deepen our understanding of this process. Finally, we are conducting single-particle analyses to confirm the structural of the liposome-coated sensors and to assess their responsiveness and stability under biologically relevant conditions."

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